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# Effect of Chemical State on the Decay Rate of $^{51}Cr$

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The chemical effect on the electron capture probability of $^{51}Cr$ is observed for the first time. The experimental result with the source combination of $Na_2CrO_4-CrCl_3$ is $\lambda(Cr^{6+})-\lambda(Cr^{3+})=(5.3\pm2.1)\times10^{-4}\lambda(Cr^{3+})$. This results is compared with a simple theoretical estimation calculated following the method of Makariūnas. The theoretical value for the relative change in the decay constant of $^{51}Cr$ between hexavalent and trivalent states is a few times greater than the experimental value. However, the present estimation gives the reasonable qualitative explanation to the experimental result.

KEY WORDS: Electron capture/ $^{51}Cr$/ Measured $T_{1/2}$/ Chemical effect/

## I. INTRODUCTION

Since the discovery of radioactivity, it has been believed that decay rate of radioactive materials is constant and can not be affected by any external condition. Therefore, the suggestion by Segrè[1] and Daudel[2] in 1947 that in the case of nuclear decay involving the orbital electrons, the decay rate might be altered by an external influence, drew a strong attention to physicists. There are two types of decay where the orbital electrons directly concern the nuclear decay, *i.e.*, the electron capture and the internal conversion.

In the case of electron capture (EC), the decay constant $\lambda$ is proportional to the electron density at the nucleus $|\psi(0)|^2$. When the electron state is perturbed by external condition (chemical or physical), a small but measurable change in $|\psi(0)|^2$ or in $\lambda$ can be expected. Nuclei of smaller atomic numbers are favorable for observation of change in $\lambda$, because the atomic shells making a dominant contribution to the capture probability are strongly influenced by environmental conditions. From this point of view, $^{7}Be$ is the most suitable nucleus. Actually, the change in $\lambda$ of $^{7}Be$ in various chemical and physical states has been measured by many workers.[3] Experimental studies for several other nucldies ($^{85}Sr$, $^{89}Zr$, $^{97}Ru$, $^{131}Ba$) have been reported.[3] The unusually large change in $\lambda$ for $^{64}Cu$ obtained by Kemény[4] has not yet been confirmed by other workers.

Only one theoretical model of the chemical effect on the decay constant for electron capture decay has been reported up to the present. Makariūnas[5] calculated the contributions of valence electrons to the total capture probabilities, and gave upper limits of the change in $\lambda$ for almost all nuclei. He suggested that the change in $\lambda$ is not simply determined by the change in the electron density at the nucleus, but the correction factor for the exchange and overlap of electron wave functions plays an important role especially in the case of light elements.

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In the present work, change in the decay constant of $^{51}Cr$, $\lambda_{Cr}$, in two ionized states, trivalent and hexavalent states, was observed for the first time. The experimental result was compared with a simple theoretical estimation calculated following the method of Makariūnas. The decay mode of $^{51}Cr$ is EC with a half-life of 27.8 d, where about 90% leads to the $^{51}V$ ground state (branch 1, transition energy $Q_1$=752 keV), and about 10% to the first excited state of $^{51}V$ (branch 2, $Q_2$= 432 keV). This excited state promptly decays to the ground state of $^{51}V$ by emitting the 320.1 keV $\gamma$ rays. Since both branches are allowed transitions, electron capture from $s$ and $p_{1/2}$ shells is predominant.[6] Alteration in the capture probability can be expected under different ionized states, because electronic-state in the inner shells, especially in 3$s$ shell, would be perturbed by the change in the valence state.

A small change in $\lambda_{Cr}$ was measured by menas of the so-called differential method. With an intention of measuring a minute difference in the intensities of two $\gamma$-ray sources by this method, we constructed a double ionization chamber, which consists of two essentially identical chambers. For measurement of ion current, a conventional sampling method was adopted. Details of the chamber construction and the measuring system were reported earlier.[7] The change in $\lambda_{Cr}$ was measured for two chemical forms ($CrCl_3$, $Na_2CrO_4$).

## II. THEORETICAL

The accurate estimation of the chemical effect on EC rate is very difficult. However, if we are interested only in the relative magnitude of $\lambda$ between different states, it is possible to make approximate estimations of this effect. In this paper, we presented rough theoretical estimations of the chemical effect on $\lambda_{Cr}$ in three ionized states; neutral, trivalent, and hexavalent states.

The transition probability of EC decay is given by[6]

$$\lambda = G^2(4\pi^2)^{-1}\sum_x q_x^2 C_x n_x \beta_x^2 B_x , \qquad (1)$$

where $G$ is the weak-interaction coupling constant; the running index x referes to the different shells of the atomic electrons from which an electron can be captured; $q_x$ is the neutrino momentum emitted when the x-shell electron is captured and corresponds to the transition energy; $C_x$ corresponds to the shape factor of $\beta$ decay, including the nuclear matrix elements; $n_x$ is equal to the relative occupation number of electrons in the x shell (for fully occupied shell, $n_x$=1); $\beta_x$ is the Coulomb amplitude of the bound-state electron radial wave function; $B_x$ is the correction factor for the overlap and exchange effects of electrons involved. In Eq. (1), the atomic units are employed, *i.e.*, $m=e=\hbar=1$ and $c$=137.037. For allowed transition as in the case of $^{51}Cr$, the shape factor $C_x$ is independent of the electronic state, and the major contribution to the total capture probability comes from $s$ electrons. Considering its large branching ratio, only branch 1 will be considered here. Then, the relative change in the decay constant of $^{51}Cr$, $\Delta\lambda/\lambda_{Cr}$, is simply expressed by

$$\Delta\lambda/\lambda_{Cr} = \sum_x \Delta A_x / \sum_x A_x , \qquad (2)$$

where x refers only to $s$ shells, *i.e.*, K, $L_1$, $M_1$, and $N_1$ shells (hereafter the subindex 1 is not shown). $A_x$ and $\Delta A_x$ denote the product $q_x^2 n_x \beta_x^2 B_x$ and its variation, respectively. The neutrino momentum is given as $q_x=(Q_1-E_{Bx})/137.037$, where $E_{Bx}$ is x-shell binding energy. The factor $n_x\beta_x^2$ is equal to x-shell electron density at the nucleus, $\omega_x|\psi_x(0)|^2$, where $\omega_x$ is the occupation number of the orbital x and $\psi_x(r)$ is its electron wave function. The correction factor $B_x$ is given by Bahcall's formulas[8] as

$$B_x = |f(x')/\psi_x(0)|^2 , \tag{3}$$

where $f(x')$ is the capture amplitude. For example, in the case of K capture, $f(1s')$ is given by

$$\begin{aligned} f(1s') = &\langle 2s'|2s\rangle\langle 3s'|3s\rangle\psi_{1s}(0) - \langle 2s'|1s\rangle\langle 3s'|3s\rangle\psi_{2s}(0) \\ &- \langle 3s'|1s\rangle\langle 2s'|2s\rangle\psi_{3s}(0) , \end{aligned} \tag{4}$$

where the primed orbital refers to the daughter atom. For other orbitals, $f(x')$ is given similarly. The first term of the right-hand side in Eq. (4) indicates the direct K capture, and the coefficient $\langle 2s'|2s\rangle\langle 3s'|3s\rangle$ becomes less than unity due to the imperfect overlap of electron wave functions. The second and the third terms are the exchange correction terms, *i.e.*, the second shows the exchange between $1s$ and $2s'$ electrons, and the third between $1s$ and $3s'$.

The calculations were made for three states, *i.e.*, neutral, trivalent, and hexavalent states. The electron configurations corresponding to these states are assumed to be $(Ar)(d)^5(4s)$ for neutral Cr, $(Ar)(3d)^3$ or $(Ar)(3d)^2(4s)$ for $Cr^{3+}$, and the same configuration as neutral Ar atom for $Cr^{6+}$. Hereafter, $(Ar)(3d)^3$ is denoted by $Cr^{3+}$A and $(Ar)(3d)^2(4s)$ by $Cr^{3+}$B. After electron capture of Cr, the electron configuration in the daughter atom, V, is the same as that of the parent atom, Cr, except for an electron hole resulting from the electron capture.

We used the Hartree-Fock-Slater (HFS) approximation. In this model, the exchange potential is given by:

$$V_{Ex}(r) = -6[3|\rho(r)|/(8\pi)]^{1/3} , \tag{5}$$

where $\rho(r)$ is the spherically averaged total electronic charge density and expressed as

$$\rho(r) = (1/4\pi)\sum_x \omega_x|\psi_x(r)|^2 . \tag{6}$$

The occupation number $\omega_x$ is determined from each electron configuration.

The calculations were carried out by the use of a modified version of a HFS program of Herman and Skillman.[9] Using the electron wave functions and the binding energies of all $s$ shells for Cr and V obtained from the HFS calculations, the factors, $E_{Bx}$, $n_x\beta_x^2$, and $B_x$ were estimated. The results are listed in Table I, where the values of $\beta_x^2$ are normalized to $\beta_K^2$ for neutral Cr. The contribution of x shell to the total capture rate can be evaluated by the product $A_x=q_x^2 n_x\beta_x^2 B_x$. The values of $A_x$ normalized to $A_K$ for neutral Cr are also included in the Table.

Table I. The responsible factors for the alteration in $\lambda_{Cr}$, $n_x$, $E_{Bx}$, $\beta_x^2$, $B_x$, and $A_x=n_x\beta_x^2B_x(Q-E_{Bx})$, for all shells for three ionized states of Cr.

| Shell | $n_x$ | $E_{Bx}$ (keV) | $\beta_x^{2*}$ | $B_x$ | $A_x^{**}$ | $\Sigma A_x$ |
|---|---|---|---|---|---|---|
| Neutral Cr [(Ar)$(3d)^5(4s)$] | | | | | | |
| K | 1 | 5.91 | 1 | 0.9876 | 1 | |
| L | 1 | 0.68 | 0.09004 | 1.1197 | 0.10352 | 1.12151 |
| M | 1 | 0.08 | 0.01268 | 1.3333 | 0.01738 | |
| N | 0.5 | 0.00 | 0.00067 | 1.7832 | 0.00061 | |
| $Cr^{3+}$A [(Ar)$(3d)^3$] | | | | | | |
| K | 1 | 5.95 | 0.99994 | 0.9885 | 1.00074 | |
| L | 1 | 0.72 | 0.09004 | 1.1227 | 0.10379 | 1.12289 |
| M | 1 | 0.12 | 0.01324 | 1.3487 | 0.01836 | |
| N | 0 | — | — | — | — | |
| $Cr^{3+}$B [(Ar)$(3d)^2(4s)$] | | | | | | |
| K | 1 | 5.96 | 0.99984 | 0.9878 | 0.99991 | |
| L | 1 | 0.73 | 0.09005 | 1.1174 | 0.10330 | 1.12326 |
| M | 1 | 0.13 | 0.01363 | 1.3062 | 0.01831 | |
| N | 0.5 | 0.04 | 0.00200 | 1.6890 | 0.00174 | |
| $Cr^{6+}$ [(Ar)] | | | | | | |
| K | 1 | 6.03 | 0.99970 | 0.9884 | 1.00019 | |
| L | 1 | 0.80 | 0.09023 | 1.1208 | 0.10380 | 1.12449 |
| M | 1 | 0.19 | 0.01483 | 1.3445 | 0.02050 | |
| N | 0 | — | — | — | — | |

* Normalized to $\beta_K^2$ for neutral Cr.
** Normalized to $A_K$ for neutral Cr.

From the values of Table I, $\Delta\lambda/\lambda_{Cr}$ was calculated by Eq. (2) for three combinations; $Cr^{3+}$A—neutral Cr, $Cr^{3+}$B—neutral Cr, and $Cr^{6+}$—neutral Cr. In order to estimate the relative contributions of factors, $q_x^2$, $n_x\beta_x^2$, and $B_x$ to $\Delta\lambda/\lambda_{Cr}$, relative changes of each shell were also calculated. The results are listed in Table II.

## III. EXPERIMENTAL

### 1. Source Preparation

The radioisotope $^{51}Cr$ is commercially available as the chemical form of $Na_2CrO_4$ in physiological salt solution. The initial activity of $^{51}Cr$ used in the present study was 10 mCi. Preliminary search showed no appreciable radioactive contaminants.

To observe the chemical effect on the decay constant of $^{51}Cr$, two ionized states of Cr, trivalent ($CrCl_3$) and hexavalent ($Na_2CrO_4$), were prepared. Each sample was produced from 5 mCi $^{51}Cr$.

As a carrier of $CrCl_3$ sample, 2.0 ml of 0.01M-$Na_2CrO_4$ solution was added to 5 mCi $^{51}Cr$. Since the ratio of carrier and $^{51}Cr$ atoms is $10^4$, the latter is expected to be fully isolated from each other, and hence the influence of $^{51}V$, the daughter

Table II. Relative differences in $A_x$, $n_x\beta_x^2$, $B_x$, and $q_x^2$ between ionized and normal states of Cr ($\times 10^{-3}$).

| Shell | $\Delta A_x/\Sigma A_x$ | $\Delta(n_x\beta_x^2)/(n_x\beta_x^2)$ | $\Delta B_x/B_x$ | $\Delta(q_x^2)/q_x^2$ |
|---|---|---|---|---|
| $Cr^{3+}$A [(Ar)$(3d)^3$]—Neutral Cr | | | | |
| K | 0.66 | −0.06 | 0.91 | −0.11 |
| L | 0.24 | 0.00 | 2.68 | −0.11 |
| M | 0.86 | 44.16 | 11.55 | −0.11 |
| N | −0.54 | −1000.00 | — | — |
| $\Delta\lambda/\lambda_{Cr}$ | 1.22 | | | |
| $Cr^{3+}$B [(Ar)$(3d)^2(4s)$]—Neutral Cr | | | | |
| K | −0.06 | −0.16 | 0.20 | −0.11 |
| L | −0.18 | 0.11 | −0.19 | −0.11 |
| M | 0.84 | 74.92 | −20.33 | −0.11 |
| N | 1.05 | 1985.07 | −52.83 | −0.11 |
| $\Delta\lambda/\lambda_{Cr}$ | 1.65 | | | |
| $Cr^{6+}$ [(Ar)]—Neutral Cr | | | | |
| K | 0.17 | −0.30 | 0.81 | −0.32 |
| L | 0.25 | 2.11 | 0.98 | −0.32 |
| M | 2.75 | 169.56 | 8.40 | −0.32 |
| N | −0.54 | −1000.00 | — | — |
| $\Delta\lambda/\lambda_{Cr}$ | 2.63 | | | |

of $^{51}Cr$, as chemical impurity can be neglected. Then 0.3 ml of 12-N HCl was added, by which the color of the solution became orange, indicating the production of $Na_2Cr_2O_7$. For reduction, 1.5 ml of acetaldehyde was added, resulting in the production of trivalent Cr ion with the color of dark green. The reduction process takes place as

$$Cr_2O_7^{2-}+8H^+ \rightarrow 2Cr^{3+}+4H_2O+5O_2$$

After the sample is dried up, the residue is still dark green, indicating the production of $CrCl_3$ crystal (ionic radius of Cr=0.76 Å).

Similarly, we prepared the $Na_2CrO_4$ sample, where 2.0 ml of 0.01M-$Na_2CrO_4$ solution was first added as a carrier to 5 mCi $^{51}Cr$. Then the solution was dried up by heating with an incandescent lamp for 5∼6 h. The color of the sample thus obtained is yellow, showing the production of $Na_2CrO_4$ crystal (ionic radius of Cr= 0.44 Å).

Both samples thus prepared are chemically stable, and do not show any deliquescence nor efflorescence. These samples were tightly sealed in a glass vial and used for the measurement of $\Delta\lambda/\lambda_{Cr}$.

## 2. Measurements of Ion Current

For the detection of a minute difference in $\lambda_{Cr}$, measurements of ion current employed in this work were made by the differential method which has been exten-

sively used in investigation of the change in $\lambda$. In this method, two radioactive sources are simultaneously measured under as nearly identical conditions as possible and only the difference current produced by two sources is measured as a function of time. The present measuring system consists of a double ionization chamber, vibrating reed electrometer (input impedence$=10^{10}$ $\Omega$), and conventional sampling system. Using this system, a minute change in intensities (1 part in $10^5$) of two $\gamma$-ray sources can be observed. Since the details of the chamber construction and the whole performance of the system were reported previously,[7] only the principle of measurements is described here.

Let $E_1(0)$ and $E_2(0)$ be the initial ion currents produced by in the chambers 1 and 2, respectively. The ion current at $t$ is expressed as $E_1(t)=E_1(0)\exp(-\lambda_1 t)$ and $E_2(t)=E_2(0)\exp(-\lambda_2 t)$, where $\lambda_1$ and $\lambda_2$ are the decay constants of two $\gamma$-ray sources mounted in chambers 1 and 2, respectively. Since the collecting voltage of both chambers are opposite in sign, the difference ion current is given by

$$\Delta E(t) = E_1(0)\exp(-\lambda_1 t) - E_2(0)\exp(-\lambda_2 t)\,. \qquad (7)$$

Supposing $\lambda_2=\lambda_1+\Delta\lambda(\Delta\lambda/\lambda_1 \ll 1)$, $\Delta E(t)$ is approximately obtained as

$$\Delta E(t)\exp(\lambda_1 t) = [E_1(0)-E_2(0)]+E_2(0)\Delta\lambda t\,. \qquad (8)$$

Since this equation has a linear form, the slope of a least-squares line of the observed values is equal to $E_2(0)\Delta\lambda$, from which one can calculate $\Delta\lambda/\lambda$. The difference in the ion currents is mesaured through a vibrating reed electrometer with the aid of a conventional sampling method, *i.e.*, for obtaining the mean value of ion current varying each moment due to the statistical nature of decay, the output ion current is recorded intermittently and the mean value in a certain time period is calculated. For ion current of $2\times10^{-10}$ A, $\sigma/E_1(0)=0.003\%$, where $\sigma$ is the standard deviation. This is the limit of accuracy of the present device.

As shown in Eq. (8), the slope of the observed value $\Delta E(t)\exp(\lambda_1 t)$ is proportional to the initial current $E_2(0)$ and $\Delta\lambda$. Therefore, a small $\Delta\lambda$ is easy to be found when $E_2(0)$ is made as large as possible. However, the initial ion current $E_2(0)$ [or $E_1(0)$] should be chosen as about $2\times10^{-10}$ A or less, because for a larger ion current the linearity of output current slightly deteriorates, probably due to the recombination of ions. In addition, it has been found that for an initial ion current larger than $2\times10^{-10}$ A, the present chamber system sometimes gives a sine-wave shape fluctuation of output current as a function of time. This phenomenon may appear in relation to the complicated structure of the chamber system. In the actual case, the initial intensities of $^{51}$Cr sources were about 5 mCi, giving ion currents of about $0.9\times10^{-10}$ A.

The measurements of $\Delta E(t)$ were carried out for 4~6 h per day and were continued for about 40 d or more. Taking into consideration the time interval for stabilization of the chamber system, the data obtained in the first 10~15 d were not adopted for analysis. During the measurements, the chamber system and subsidiary electric instruments were kept from any disturbance. The temperature in the room was carefully controlled and was kept at $20.0\pm0.1$°C.

## IV. RESULTS AND DISCUSSION

With a source pair, $Na_2CrO_4(Cr^{6+})$–$CrCl_3(Cr^{3+})$, the chemical effect on $\lambda_{Cr}$ was measured. As a reference, the measurements with two chemically identical sources ($Na_2CrO_4$) were also carried out. In Fig. 1, the experimental points and a least-squares line for each combination are shown.

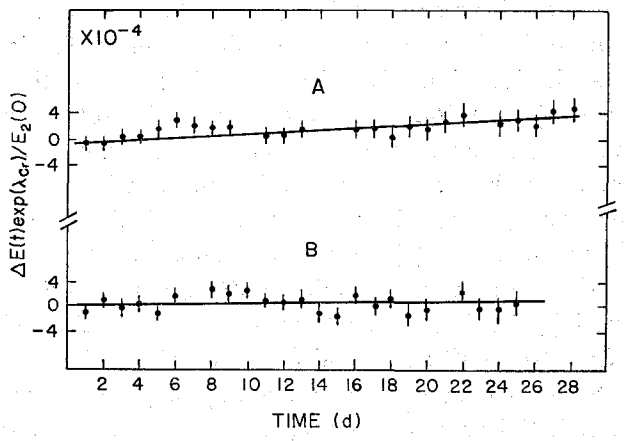


Fig. 1. Two combinations of $^{51}Cr$ sources are measured. Data are normalized to the initial output current $E_2(0)$ after subtracting the initial difference current. (A) $CrCl_3$—$Na_2CrO_4$; (B) $Na_2CrO_4$—$Na_2CrO_4$.

As shown in the figure, the fluctuation width of the experimental points is considerably large, and hence the value of $\chi^2$ for each curve becomes 3~4, indicating that the systematic error can not be neglected. The most responsible factor for this systematic error is the instability of the measuring device, especially of the chamber system. In the figure, however, a slope appears for the source pair, $Cr^{6+}$–$Cr^{3+}$, while the reference curve is horizontal within the experimental error. Since the slope of the reference curve is equivalent to $\Delta\lambda/\lambda_{Cr}=(0.4\pm1.9)\times10^{-4}$, we can expect that the chemical effect on $\lambda_{Cr}$ is measured with the acuracy of $2\times10^{-4}$. The values of $\Delta\lambda/\lambda_{Cr}$ obtained from the slope of $Cr^{6+}$–$Cr^{3+}$ pair is

$$\Delta\lambda/\lambda_{Cr} = (5.3\pm2.1)\times10^{-4},$$

where $\Delta\lambda=\lambda_{Cr}(Cr^{6+})-\lambda_{Cr}(Cr^{3+})$. This result shows the chemical shift in $\lambda_{Cr}$, and this is the first experimental evidence for the chemical effect on $\lambda_{Cr}$.

According to the theoretical estimation of $\Delta\lambda/\lambda_{Cr}$ in Chapter II, the expected relative change in $\lambda_{Cr}$ between hexavalent and trivalent states is $9.8\sim14.1\times10^{-4}$ (see Table II), being a few times larger than the experimental result. However, this theoretical estimation can give a qualitative explanation to the experiments.

As shown in the Table, $\lambda_{Cr}$ in ionized states is slightly ($\sim10^{-3}$) larger than that in neutral state mainly due to the increase in M-shell electron density at the nucleus. This dominant contribution of M shell is attributed to the so-called squeezing effect on the 3*s*-electron wave function. A typical squeezing effect is seen in $Cr^{6+}$, where the valence electrons are completely removed. The absence of a 4*s* electron de-

creases $\lambda_{Cr}(Cr^{6+})$ by about 0.05%, while the squeezing in $3s$ electrons increases it by about 0.3%. A similar situation in the N and M shells is also found for $Cr^{3+}A$ $[(Ar)(3d)^3]$, where a half of the valence electrons are removed. In this case, the magnitude of the squeezing of $3s$ electron is about a quarter of that for $Cr^{6+}$, and the increase in $\lambda_{Cr}(Cr^{3+}A)$ is about a half of that in $\lambda_{Cr}(Cr^{6+})$. In $Cr^{3+}B[(Ar)(3d)^2(4s)]$, however, both N- and M-shell contributions increase $\lambda_{Cr}(Cr^{3+}B)$ by about 0.1% because $4s$ electron is still involved in $Cr^{3+}$ ion. This increase in the N-shell contribution comes from the decrease in the screening potential for $4s$ electron, which is caused by delocalization of three $3d$ electrons. In any case, the contributions of inner shells (K and L) to $\Delta\lambda/\lambda_{Cr}$ are generally not so important except for $Cr^{3+}A$, in which relatively large contribution of K shell to the change in $\lambda_{Cr}$ come from $\Delta B_K$. This is probably due to the decrease in exchange integrals between $4s'$ (in V) and other $s$ electrons (in Cr). It is also found from Table II that the contribution of the factors $q_x^2$ and $B_x$ in Eq. (1) are not so important.

Concerning the contribution of $\Delta B_x$ to $\Delta\lambda$, Makariūnas[5] suggested its important role, especially for light elements such as $^7$Be. He also indicated that two models for $B_x$, *i.e.*, Bahcall's[8] and Vatai's[10] ansatz give different contributions of the valence electrons to total capture probability $\lambda_{val}$. According to him, the former gives about 40% larger value for $\lambda_{val}$ than the latter. Therefore, the present theoretical estimation may be reduced to some extent if we adopted Vatai's formula, in which the overlap and exchange between $s'$ and $p$ or $d$ are also taken into account.

Uncertainty of the present estimation mostly comes from the fact that the simplest ground-state electron configuration is assigned to each ionized state. In the actual compounds, such a simple configuration would not be expected, and the interaction between Cr and the surrounding elements can not be neglected as well. H. -Reinecke *et al.*[11] compared the measured $\Delta\lambda/\lambda$ for $^{97}$Ru to the values obtained by relativistic self-consistent-field calculation for free ion, and found a fairly good agreement. Although this agreement does not prove that the free-ion approximation is reliable, it can be expected that the present theoretical model gives a good approximation for estimation of $\Delta\lambda/\lambda_{Cr}$.

For further discussion, more realistic model is required. In addition, more experimental data with improved accuracy should be required to get more detailed informations. If we can measure a change in $\lambda_{Cr}$ between neutral state (metallic state) and ionized state, the test for the theoretical model will be made with more reliability.

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